Optical Measurements of Soot and Temperature Profiles in Premixed Propane-Oxygen Flames

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ABSTRACT

Two laser diagnostic techniques were used to measure soot volume fractions, number densities and soot particle radii in premixed propane/oxygen flat flames. The two techniques used were two-wavelength extinction, using 514.5 nm-632.8 nm and 457.9 nm-632.8 nm wavelength combinations, and extinction/scattering using 514.5 nm light. The flames were fuel-rich (equivalence ratios from 2.1 to 2.8) and had cold gas velocities varying from 3.4 to 5.5 cm/s. Measurements were made at various heights above the sintered-bronze, water-cooled flat flame burner with the equivalence ratio and cold gas velocity fixed. Also, measurements were made at a fixed height above the burner and fixed cold gas velocity while varying the equivalence ratio. Both laser techniques are based on the same underlying assumptions of particle size distribution and soot optical properties. Full Mie theory was used to determine the extinction coefficients, Kext, and the scattering efficiencies, Qvv. Temperature measurements in the flames were made using infra-red radiometry.

Good agreement between the two techniques in terms of soot particle radii, number density and volume fraction was found for intensity ratios (I/I_0) between 0.1 and 0.8. For intensity ratios higher, or lower than this range, the differences in extinction coefficients at the wavelengths chosen for the two-wavelength method are too small to give accurate results for comparing particle radii and number densities. However, when comparing only soot volume fractions, the agreement between the two techniques continued to be good for intensity ratios up to 0.95.

Introduction

Many studies, such as those done by Bockhorne et al. (1981), Prado et al. (1981), and D'Allesio et al. (1977) can be found on the formation of soot in premixed flames. There are also several review

articles by Wagner (1981), Haynes and Wagner (1981), and Baumgartner et al. (1984). The objective of this study was to compare two laser diagnostic techniques by measuring soot volume fractions, soot particle radii and number densities in premixed propane/oxygen flat flames. Measurements were made at various heights above the sintered bronze, water-cooled burner while fixing the equivalence ratio and cold gas flow rate. Also, measurements were made at a fixed height above the burner (15 mm) and fixed cold gas flow rate while varying the equivalence ratio. Temperature measurements were made using infra-red radiometry. two laser techniques chosen for comparison were the technique, scattering/extinction used by researchers, including Santoro et al. (1983)and Dobbins and Mulholland (1984), and the two-wavelength extinction method used by Bard and Pagni (1979, 1981a and b, 1986) and by Beier and Pagni (1981, 1983). laminar flat flame provided a well-known, stable, easily-controlled environment in which to compare the two techniques.

Experimental Techniques

Scattering/Extinction Technique

A 2W argon ion laser was the source of the 514.5 the used for scattering/extinction The overall optical system is shown in measurements. Fig. 1. A polarization rotator was used to vertically polarize the light which was then focused into the test section. After the test section, the light was collected by a lens and focused onto a 514.5±10 nm line filter and neutral density filter (ND=3) placed in front of a photodiode. The scattered light at 90 degrees to the beam path was measured by a separate photomultiplier/lock-in amplifier system. scattered light detecting system consisted of a 10-mm diameter light stop placed just before a focusing lens, a polarization analyzer, a 1-mm aperture and a narrow bandwidth (514.5±1 nm) line filter placed on the end of a 15-cm-long black cylinder on the front of an endwindow photomultiplier tube. The scattering system was set up to have unity magnification of the test section. The photomultiplier output was processed through a lock-in amplifier to rid the signal of all but the laser light. The lock-in amplifier output and the output from the photodiode readout were sent to an analog-digital converter in a micro-computer. output of each of these detectors was then recorded 400 times in 20 seconds and the mean values of these readings were used in the data analysis.

The system was calibrated after each data point using nitrogen, with a known scattering cross section (Rudder and Bach, 1968), $\sigma_{\rm O}$, and number density, N_O, and assuming negligible absorption. The scattering efficiency for soot can then be determined from:

$$Q_{\text{vv,soot}} = \left(\sigma_{\text{o}} N_{\text{o}} \right)_{N_{2}} \frac{I_{\text{s,soot}}}{I_{\text{s,N}_{2}}} \left(\frac{I(L)}{I_{\text{o}}} \right)^{-(\ell+d)/L}$$
(1)

where I_s is the measured scattered light, I_o is the incoming laser light intensity, measured by the photodiode with no flame present, I(L) is the light intensity after passing a distance L through the flame. The location of the probe volume in the flame determines ℓ and d, see Fig. 2. For sampling in the center of the flame, $\ell=d=L/2$.

Two-Wavelength Technique

The optical system for the two-wavelength technique is the same as the extinction portion of the scattering/extinction technique except wavelengths of light were used. Only two wavelengths are needed for the two-wavelength technique, except when the normalized extinction coefficient, X_{ij} (defined later) is greater than 1. Then there are two possible values for R_{max} and a second combination of wavelengths is necessary to determine the correct R_{max} . The wavelength combinations used were 457.9 nm-632.8 nm and 514.5 nm-632.8 nm. The 457.9 and 514.5 nm lines were supplied by the argon ion laser and the 632.8 nm line was supplied by a 15-mW helium neon laser. A cube beamsplitter was used to combine the light from the two lasers so they passed through the same path in the Two equilateral prisms were used after the collecting lens to separate the individual wavelengths. Laser line filters for each of the three wavelengths were placed in front of the each of the three corresponding detectors. The laser beam pathlength through the flame, L, was measured for each data point using a cathetometer. The extinction coefficient for each wavelength can then be experimentally determined from:

$$K_{\text{ext}} = -\frac{1}{L} \ln \left(\frac{I(L)}{I_0} \right)$$
 (2)

Analysis

The two values determined experimentally are the extinction coefficient, K_{ext} , and the scattering efficiency, Q_{vv} . The extinction coefficient is related to the extinction efficiency, Q_{ext} , of each particle of radius, R, and to the particle number density, N(R), by

$$K_{\text{ext}} = \int_{0}^{\infty} N(R) Q_{\text{ext}}(\lambda, m, R) \pi R^{2} dR$$
 (3)

where λ is the wavelength of the light source and $m=n(\lambda)-ik(\lambda)$ is the refractive index of soot in this case (Pagni and Bard, 1979). Assuming spherical particles, $Q_{\text{ext}}(\lambda,m,R)$ is found from Mie scattering theory (Mie, 1908, Kerker, 1969, and Hottel and Sarofim, 1967). The scattering efficiency, Q_{VV} , is related to the particle scattering cross-section, $C_{\text{VV}}(\theta,\lambda,R,m)$ by

$$Q_{VV} = \int_{0}^{\infty} N(R) C_{VV}(\theta, \lambda, R, m) dR$$
 (4)

where θ is the scattering angle (90 degrees in this case). Again, Mie scattering theory provides a value for C_{---}

for C_{VV} A Gamma distribution (Hahn and Shapiro, 1968) with the constraint of a ratio of standard deviation to mean particle radius, $\sigma/r_{\rm m}=1/2$, was chosen based on size measurements in premixed flames (Wersborg et al., 1973). In terms of the most probable radius, $R_{\rm max}$, and the total particle concentration, $N_{\rm O}$, the size distribution is

$$N(R) = N_{O} \left(\frac{27 R^{3}}{2 R^{4}_{max}} \right) exp\left(\frac{-3 R}{R_{max}} \right)$$
 (5)

Substituting Eq.(5) into Eq.(3) yields an equation for K_{ext} in terms of the two unknowns N_{O} and R_{max} . The other equation needed to determine these two parameters is provided either by extinction measurements at another wavelength (two-wavelength technique) or by scattering measurements at the same wavelength (scattering/extinction technique).

Two-Wavelength Technique

Values for K_{ext} can be obtained by integrating Eq.(3) numerically at fixed wavelengths 457.9 nm, 514.5 nm and 632.8 nm for various values of R_{max} . These extinction coefficients can then be presented as non-

dimensional extinction coefficients defined as (Bard and Pagni, 1981a):

$$\tau'(\alpha_{\max}, \lambda, m) = \frac{K_{\text{ext}}(\alpha_{\max}, \lambda, m)}{N_{\text{o}} R_{\text{max}}^{2}}$$
(6)

where α_{max} is the characteristic size parameter, $2\pi R_{\text{max}}/\lambda$. If $\alpha_{\text{max}}<<1$, the absorption limit applies where K_{ext} is independent of the size distribution and only depends on the optical properties of the particles. Then the ratio of K_{ext} 's at two different wavelengths is:

$$\left[\begin{array}{c} \frac{K_{\text{ext}}(\lambda_{i})}{K_{\text{ext}}(\lambda_{j})} \right] = \left[\begin{array}{c} K_{i} \\ \hline K_{j} \end{array}\right]_{\text{abs}} = \frac{\lambda_{j} F_{a}(\lambda_{i})}{\lambda_{i} F_{a}(\lambda_{j})}$$
(7)

where

$$F_{a}(\lambda) = \frac{n^{2} k}{\left[n^{2} - (nk)^{2} + 2\right]^{2} + 4n^{4}k^{2}}$$
 (8)

The refractive indices used were (Lee and Tien, 1981 and 1982): 1) n=1.94, k=0.58 for λ =457.9 nm; 2) n=1.93, k=0.52 for λ =514.5 nm; and 3) n=1.89, k=0.48 for λ =632.8 nm.

In the large particle limit, $\alpha_{\text{max}} >> 1$, $Q_{\text{ext}} \rightarrow 2$ for all λ , so $K_{i}/K_{j} \rightarrow 1$. In order to incorporate these limits into a useful parameter, Pagni and Bard (1979) suggested a normalized extinction coefficient defined as:

$$X_{ij} = \frac{\begin{bmatrix} K_i / K_j - 1 \end{bmatrix}}{\begin{bmatrix} K_i / K_j - 1 \end{bmatrix}} abs$$
 (9)

The extinction coefficient is found experimentally from Eq.(2). When the two laser beams are superimposed, the pathlength $L_i=L_j=L$, and the ratio of extinction coefficients is:

$$\frac{K_{\underline{i}}}{K_{\underline{j}}} = \frac{\left[\ln(I/I_0)\right]_{\underline{i}}}{\left[\ln(I/I_0)\right]_{\underline{j}}}$$
(10)

The measured light intensity ratios at each wavelength are used in Eq.(10). Using Eq.(10) in the numerator and Eqs. (7) and (8) in the denominator of Eq. (9) gives X_{ij} . This experimental value of X_{ij} is then used to find the corresponding R_{max} in a table of calculated values of X_{ij} and R_{max} . Using either K_{ext} and the

corresponding τ' , N_0 can be determined from Eq.(6).

Scattering/Extinction Technique

Substituting Eq.(5) into Eq.(4) gives a second equation involving N_O and R_{max} . The total particle concentration, N_O , can then be eliminated by dividing this equation for Q_{VV} by K_{ext} from Eq.(3) to give:

$$\frac{Q_{\text{VV}}}{K_{\text{ext}}} = \frac{\int_{0}^{\infty} \frac{R^{3}}{R_{\text{max}}^{4}} \exp\left(\frac{-3R}{R_{\text{max}}}\right) C_{\text{VV}} dR}{\int_{0}^{\infty} \frac{R^{3}}{R_{\text{max}}^{4}} \exp\left(\frac{-3R}{R_{\text{max}}}\right) Q_{\text{ext}} \pi R^{2} dR} \tag{11}$$

Equation (11) is then solved numerically (Bohren and Huffman, 1983, and Hildebrand, 1956) for the ratio $Q_{\rm VV}/K_{\rm ext}$ as a function of $R_{\rm max}$ which produces a table of theoretical values. With the experimental $Q_{\rm VV}/K_{\rm ext}$, determined from Eqs. (1) and (2), this table can be used to find the corresponding $R_{\rm max}$. The number density, $N_{\rm O}$, is then obtained by substituting Eq.(5) into Eq.(4) and using the result of the integration of the numerator of Eq.(11), call it β , and $Q_{\rm VV}$ from Eq.(1):

$$N_{O} = \frac{2 Q_{VV}}{27 \beta} \tag{12}$$

Soot Volume Fractions

The soot volume fraction for both techniques is given by:

$$f_{V} = \frac{4\pi}{3} \int_{0}^{\infty} R^{3} N(R) dR \qquad (13)$$

Substituting Eq.(5) into Eq.(13) and integrating gives:

$$f_v = \frac{2\pi \Gamma(7)}{3^5} N_o R_{max}^3 = 18.62 N_o R_{max}^3$$
 (14)

Experimental Results

In order to compare results with previous studies, two experimental conditions were chosen to be the same as those studied by Prado, et al., 1981, in a similar burner. These conditions were cold gas flow velocities of 3.92 and 5.5 cm/s at an equivalence ratio of 2.5. Soot volume fractions, f_V , particle radii, R_{max} , and number densities, N_O , were measured at various heights above the burner for these conditions and for a velocity of 4.71 cm/s. Also, at a fixed height above the burner of 15 mm, the equivalence ratio was varied from 2.1 to 2.8, while keeping the cold gas velocity fixed for each of the three chosen velocities. Measurements were made simultaneously using the two-wavelength method and the scattering/extinction method.

f_v, R_{max}, and N_o obtained from scattering/extinction measurements at various heights above the burner are shown in Fig.3 for the three cold gas velocities and an equivalence ratio of 2.5. soot particle sizes and number densities compare reasonably with Prado's, et al. (1981) results, when he also assumed spherical particles. However, the agglomerated particles that Prado measured with sampling probes were larger in diameter and lower in number density. Prado's results are not shown on Fig. 3 due to the use of different refractive indices in these two studies. Prado's results used Dalzell and Sarofim's (1969) value of m=1.57-i 0.56. This would lead to a larger expected soot size than for the refractive index recommended by Lee and Tien (1981, 1982) of m=1.93-i 0.52 used here. This smaller particle size leads to a slightly smaller volume fraction than Prado's results. However, similar trends in the relative amounts of soot at a particular height above the burner and cold gas flow rate are seen in both studies. At small heights above the burner (below 10 mm), the soot forms quickly, starting with a large number (over $10^{12}/\text{cc}$) of very small (less than 2 nm radius) particles. In the region from 2 to 5 mm, the sharply decreasing number density and increasing particle size produced a sharply increasing volume fraction. Near a height above the burner of 10 mm, the number of soot particles approaches a value near 10¹⁰/cc, and remains at this value for heights above 10 mm. The particle size continues to grow above this height, although at a reduced rate. The volume fraction reflects the effect of increased particle size with a relatively constant number of particles as seen by the slightly increasing volume fraction above 10 mm. The effect of cold flow velocity is negligible on the number density. However, the higher flow rates produced smaller particles and lower volume fractions for each height above the burner, reflecting shorter residence times and higher flame temperatures.

The scattering/extinction technique was used to measure the effect of varying the equivalence ratio at the three cold flow velocities, shown in Fig. 4. As expected, for increased equivalence ratios, the size

and number of soot particles increases, producing an increase in volume fraction. The effect of cold gas velocity again decreases the size and volume fraction of soot particles for increased velocities. There is little effect of the cold gas velocity on the number density.

Comparison of Two Methods

The scattering/extinction method and the two-wavelength method are compared in a flame having an equivalence ratio of 2.5 and a cold gas velocity of 3.92 cm/s. These conditions provided sufficient amounts of soot to produce strong signals for both the scattering and extinction measurements.

Soot particle size, number density and volume fractions as predicted by both the scattering-extinction and the two-wavelength techniques are presented in Fig. 5 as a function of height above the burner. There is some discrepancy below a height above the burner of 10 mm when measuring size and number density. The two-wavelength technique appears less sensitive to the change in particle sizes for the small (less than 10 nm radius) particles near the burner surface (below 10 mm) and so predicts the same size (or even slightly larger particles) here. Since this particle size is used to determine the number density, the number density predicted by the two-wavelength technique is lower than the scattering technique for measurements below the 10 mm height above the burner. However, there is excellent agreement between the two techniques when measuring volume fraction since the over-prediction of particle size by the two-wavelength method is compensated by its under-prediction of number density. It is interesting to note that the agreement between the two methods for size and number density was best for soot volume fractions above 1x10⁻⁶.

A comparison of the two techniques was made as a function of equivalence ratio at a height above the burner of 15 mm for the same (3.92 cm/s) cold gas velocity. Figure 6 shows the results for the soot particle size, number density and volume fraction for equivalence ratios from 2.1 to 2.8. Below equivalence ratio of 2.1, there was too little soot to repeatable measurements allow scattering/extinction technique. The two-wavelength method calculated impossible (negative or too-large positive) values for X_{ij}, resulting in no measurements until an equivalence ratio of 2.3. Below this equivalence ratio, the I/I_o (extinction measurement) Below this values for the three wavelengths used were nearly 1.0. Measurement of the slight variations from this value was difficult due to a small signal to noise ratio. was not evident problem as This

scattering/extinction measurements since the scattering intensity varies by a much greater amount for small changes in equivalence ratio (for example from 2.11 to 2.17, the scattering intensity doubles). The extinction measurement plays a lesser role in the final results for the scattering/extinction method. For longer pathlengths (larger burner), or more densely sooting flames, the extinction measurement would be more accurate.

The agreement between the two techniques begins to be good near an equivalence ratio of 2.5 (where the soot volume fraction exceeds 10⁻⁶). The two techniques begin to disagree slightly in the very rich (near equivalence ratio 2.8) flames. For the very sooty flames, a change in the equivalence ratio will produce a smaller relative change in the extinction intensity than in the scattering intensity. The differences between the two techniques in prediction of soot particle size and number density for the very rich flames are fairly small. There is excellent agreement in the prediction of soot volume fractions by both techniques where measurements could be taken. At equivalence ratios higher than 2.8, the flame was beginning to be unsteady, (Gaydon and Wolfhard, 1970) making it difficult to measure L.

Temperature Measurement

The flame temperature was measured using infrared radiometry (Schmidt, 1909, Tourin, 1962). Figure 7a shows the flame temperature at a height above the burner of 15 mm for cold gas velocities of 6, 9 and 12 The temperature increases with increasing flow velocity. The 9 and 12 cm/s profiles show a dip in temperature approaching stoichiometric conditions. This is the result of the increasing flame speed causing the flame to more closely approach the burner surface and allow greater heat loss to the water-cooled surface. After passing the stoichiometric equivalence ratio, the flame speed again slows down and the heat transfer to the surface decreases, seen as an increase in flame temperature.

The flame temperature as a function of equivalence ratio for a sooting flame is shown in Fig.7b. The soot volume fraction at a height above the burner of 15 mm and cold gas velocity of 3.4 cm/s was measured by scattering/extinction. There is a smooth decrease in temperature as the increasing soot radiates heat out of the flame.

Conclusion

Soot volume fractions, particle sizes and number were measured in various premixed densities propane/oxygen flames by two laser techniques. The two methods compared favorably in flames where the extinction intensity ratios (I/Io) were between 0.1 and 0.8. Within this range, the two techniques produced good agreement in prediction of $R_{\text{max}},\ N_{\text{O}},\ \text{and}\ f_{\text{V}}.$ The two methods agreed in the prediction of f_{V} alone for intensity ratios up to 0.95. There is always a refractive indices of soot question of which (Charalampopoulos and Felske, 1987) should be used, and which size distribution and particle shape is correct. The good agreement between the two techniques affirms the size distribution and optical properties chosen.

Flame temperature was successfully measured using infra-red radiometry in both a sooting and a non-sooting flame.

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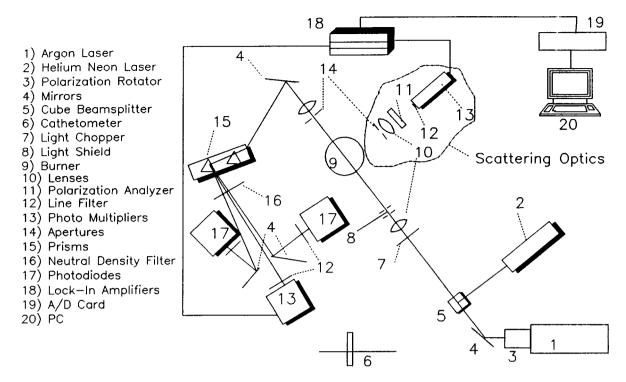


Figure 1: Schematic of Experimental Set-Up for Two-Wavelength and Scattering/Extinction Optical Diagnostic Techniques

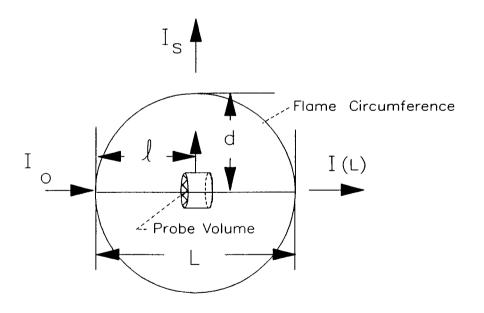
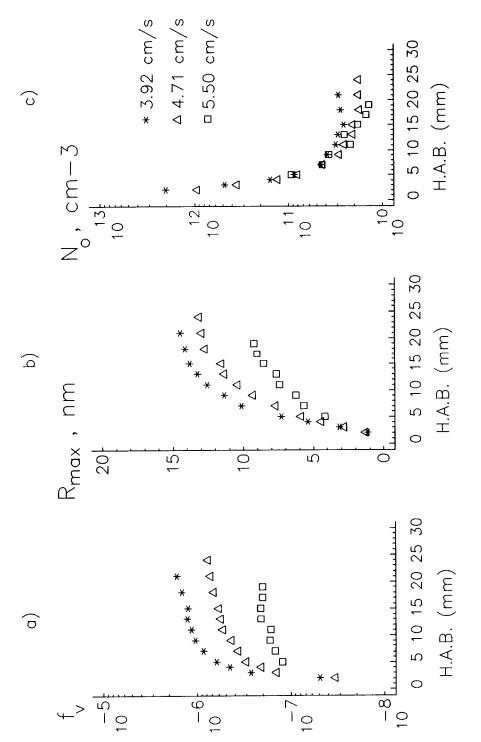


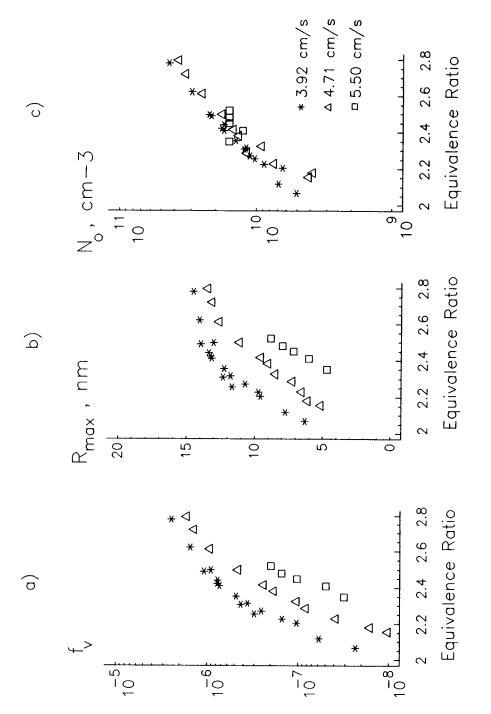
Figure 2: Top view of probe volume in the cylindrical flame. I_0 is the incoming laser intensity; I(L) is the intensity after passing a distance L through the flame; I_g is the intensity of the scattered light; 1 is the distance to the probe volume along the incoming beam path; 1 is the perpendicular distance from the beam path to the edge of the flame.



function of height above burner (H.A.B., mm). Fuel equivalence ratio = 2.5. Cold gas a) Soot volume fraction as a velocity = 3.92, 4.71, 5.50 cm/s; b) Most probable soot particle radius (nm) as a function of H.A.B.; c) Soot number density $(1/\mathrm{cm}^3)$ as function of H.A.B. Figure 3: Scattering/Extinction Technique Results:

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a) Soot volume fraction as a function of equivalence ratio. Height above burner = 15 mm. Cold gas velocity = 3.92, 4.71, 5.50 cm/s; b) Most probable soot particle radius (nm) as a function of equivalence ratio; c) soot number density $(1/\text{cm}^3)$ as a function of equivalence ratio. Figure 4: Scattering/Extinction Technique Results:

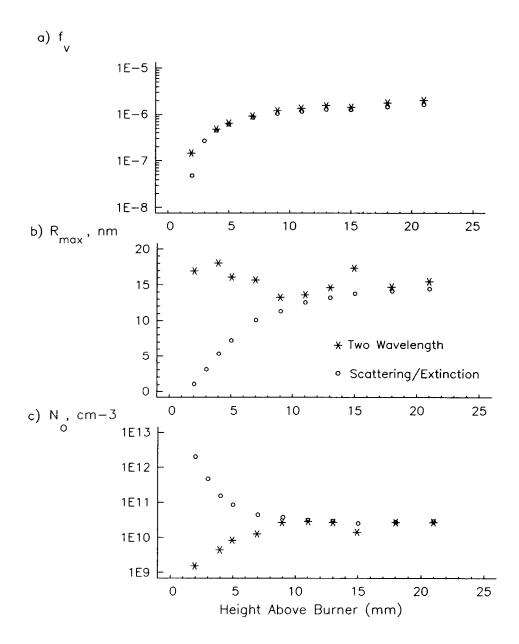


Figure 5: Comparison of Results from Two-Wavelength Method (457.9nm-632.8nm) and Scattering/Extinction: a) Soot volume fraction as a function of height above burner (mm). Equivalence ratio = 2.5, cold gas velocity = 3.92 cm/s; b) Most probable radius (nm) as a function of H.A.B.; c) Number density (1/cm³) as a function of equivalence ratio.

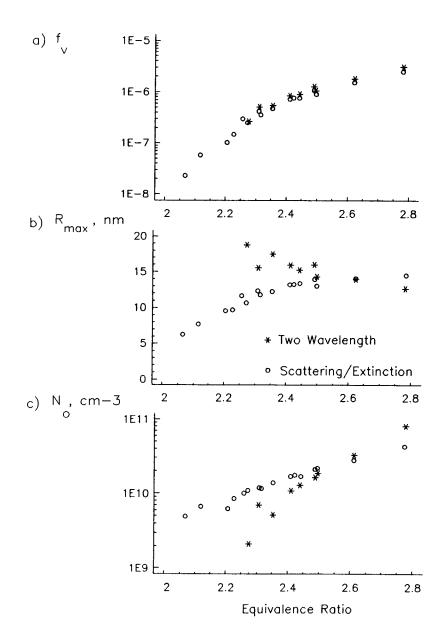
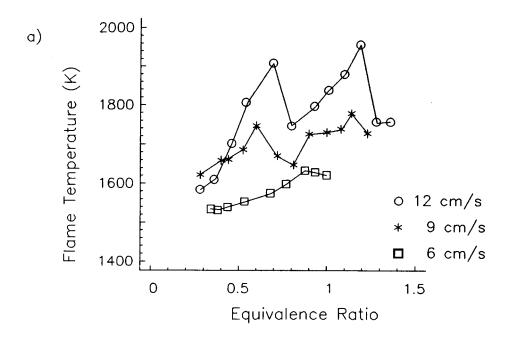


Figure 6: Comparison of Results from Two-Wavelength Method (457.9nm-632.8nm) and Scattering/Extinction for: a) Soot volume fraction as a function of equivalence ratio. H.A.B. = 15 mm, cold gas velocity = 3.92 cm/s; b) Most probable radius (nm) as a function of equivalence ratio; c) Number density $(1/\text{cm}^3)$ as a function of equivalence ratio.



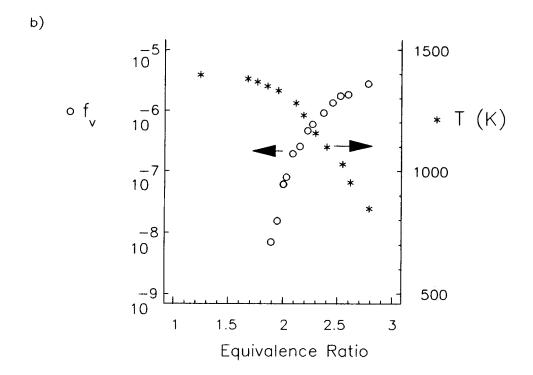


Figure 7: a) Flame Temperature as a Function of Equivalence Ratio for Cold Gas Velocities of 6, 9 and 12 cm/s at a Height Above Burner (H.A.B.) of 15 mm.; b) Flame Temperature and Soot Volume Fraction as a Function of Equivalence Ratio for a Cold Gas Velocity of 3.4 cm/s and H.A.B. of 15 mm.

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Prepared for the 1988 Winter A Illinois, November 28—Decemb University of California, Berkel 16. Abstract Two laser diagnostic techniques radii in premixed propane/oxyge 514.5 nm-632.8 nm and 457.9 n light. The flames were fuel-rich to 5.5 cm/s. Measurements were burner with the equivalence ratic above the burner and fixed cold on the same underlying assumpt used to determine the extinction ments in the flames were made of soot particle radii, number de 0.8. For intensity ratios higher, wavelengths chosen for the two-radii and number densities. How two techniques continued to be getting the continued to be getting the continued to the same underlying assumpt used to determine the extinction ments in the flames were made of soot particle radii, number de of soot particle radii and number de of soot pa	were used to measure some flat flames. The two tenter of the flames. The two tenter of the flames are ratios from the made at various heights to and cold gas velocity fing gas velocity while varying ions of particle size district coefficients, Kext, and the using infra-red radiometry ensity and volume fraction or lower than this range, wavelength method are to vever, when comparing on	ons, NASA Lewis on volume fraction chniques used we combinations, and 2.1 to 2.8) and he above the sintere ced. Also, measur g the equivalence oution and soot of e scattering efficie . Good agreemen was found for in the differences in o small to give a ly soot volume fr	as, number densities as re two-wavelength exite extinction/scattering ad cold gas velocities d-bronze, water-coole rements were made at ratio. Both laser technical properties. Full encies, Q _{vv} . Temperate the two technical properties is the two technical properties at between the two technical properties (I/I _o) be a extinction coefficient courate results for contractions.	and soot particle tinction, using using 514.5 nm varying from 3.4 d flat flame a fixed height eniques are based. Mie theory was ture measure-thniques in terms between 0.1 and its at the imparing particle
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